

# REPORT DOCUMENTATION PAGE

Form Approved  
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE 2-97	3. REPORT TYPE AND DATES COVERED FINAL REPORT 01 Sep 92 - 31 Aug 96	
4. TITLE AND SUBTITLE Thin Film Multilayer High Tc Superconductor Structures			5. FUNDING NUMBERS  61102F 2300/HS	
6. AUTHOR(S)  Professor Grover L. Larkins, Jr				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Electrical Engineering Department Florida International University Miami, Florida 33199				
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NE 110 Duncan Avenue Suite B115 Bolling AFB DC 20332-8050			10. SPONSORING/MONITORING AGENCY REPORT NUMBER  F49620-92-J-0492	
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION/AVAILABILITY STATEMENT  APPROVED FOR PUBLIC RELEASE: DISTRIBUTION UNLIMITED			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words)  Room Temperature Poling at + 10Volts (positive on the silver top electrode) of BTO in Bi-layer Structures led to changes in the Resistance vs. Temperature Curves of the 123 of the same sort as Poling above the Curie Temperature.				
14. SUBJECT TERMS			15. NUMBER OF PAGES	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED			18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED
20. LIMITATION OF ABSTRACT				

AFOSR-TR-96

97

0109

19970228 102

**Final Report  
(Technical)  
Grant No. F49620-92-J-0492DEF**

**"Thin Film Multilayer High T<sub>c</sub> Structures  
with  
Enhanced Superconductive Properties"**

**Dr. G.L. Larkins  
Florida International University  
Electrical and Computer Engineering Dept.  
ECS-314  
Miami, Fla. 33199**

**DTIC QUALITY INSPECTED 5**

## **Contents**

### **I. Summary of Overall Results:**

- a) Room Temperature Poling at +10Volts (positive on the silver top electrode) of BTO in Bi-layer Structures led to changes in the Resistance vs. Temperature Curves of the 123 of the same sort as Poling above the Curie Temperature.

### **II. Suggestions for Future Work**

- a) Elimination of "Boulder" (surface roughness) Problem common to Laser Ablated Films.
- b) Continuation with thin Four-Layer Symmetric Structure once surface roughness problems are resolved.

## I. Summary

### a) Growth Parameters

Epitaxy has been achieved with 123 deposition parameters of 790C in 200 millitorr of flowing  $O_2$  and BTO deposition at 700C in 100 millitorr of flowing  $O_2$ . Note: All deposition temperatures quoted were measured with an optical pyrometer. The resultant bi-layers showed good epitaxy with clean x-ray diffraction spectra and superconductive transitions which were approximately 1K to 2K wide with an onset at 89K to 90K and  $T_{c(\text{zero resistance})}$  of 87K to 89K for un-poled (virgin) samples.

Deposition of the BTO at temperatures in excess of about 725C resulted in "dirty" x-ray spectra with an unexplained peak at 36.5 degrees.  $T_c$ s of these films were considerably reduced from the conditions described above and often possessed a "two-step" transition which was quite broad beginning at 89K and ending at about 60K. This was taken to be indicative of the presence of non-stoichiometric phases (mixed phase or oxygen deficient material).

### b) Poling of BTO in the Bi-Layer Structure

#### Year One:

In an attempt to characterize the bi-layer a silver epoxy top electrode was painted on over the BTO and was used to pole the BTO. Since the curie temperature for BTO is 130C the poling was done at 150C. The voltage was applied throughout the 10-30 minute temperature dwell at 150C and the subsequent cool-down to room temperature. No apparent shift in the x-ray diffraction spectra was observed in a before and after poling comparison.

The resistivity vs. temperature curves did exhibit a marked change after poling of the BTO with the silver epoxy electrode positive with respect to the 123 (positive poling) but no change was observed for the opposite polarity. For positive poling the  $T_c$  onsets shifted up by an amount of from 0.5K to 1.5K and the formation of a "foot" on the R vs. T curve was often, but not always, observed (figure 1). The  $T_{c(\text{zero resistance})}$  point was not observed to shift appreciably.

The poled sample showed no change in its properties over a period of days but could be de-poled by heating to 150C for 30-60 minutes in the absence of an applied potential. The R vs. T curve of the de-poled material then returned to its virgin state (ie. no foot, onset reduced back to the original value etc.)(figure 2). Re-poling led to results ( $T_{c(\text{ONSETS})}$ ) similar to when the virgin material was poled (figure 3). A comparison of the poled virgin and the re-poled material is shown in figure 4, aside from the, in this particular case, less pronounced "foot" on the re-poled curve the two are quite similar suggesting that the effect is reversible.

#### Year Two:

We have smoothed the films down a bit but they are still rougher than desired; +/- 30nm surface roughness. Poling at elevated potentials (+10 Volts) and at Room Temperature (~20C) for 10 sec or less led to (a) large numbers of samples being destroyed due to catastrophic breakdown of the BTO in the high fields ( $10^7$  to  $10^8$  V/m) and (b) to samples which having not

broken down exhibited precisely the same behavior in the R vs T curves as those which were poled at  $T > T_{\text{Curie}}$ .

#### **Year Three:**

Results obtained are similar to those in year two, surface roughness continues to be a problem. Facility at FIU is on line for Laser Ablation but results are the same as at Xerox PARC (year two).

#### **a) Publication Status**

A paper describing the above observations of year one was presented as a poster at the European Conference on Applied Superconductivity in Goettingen Germany in early October 1993. This paper is now published (3). A paper detailing the advances and implications of the reduced temperature poling results was presented at the 1994 Applied Superconductivity Conference (4). A Master's Thesis on this topic was published in Dec. 1996 (5)

### **II. Summary of Suggested Future Work**

#### **a) Surface Roughness**

The roughness of the surfaces of the laser ablated films continues to be a problem as there is often a pinhole problem and the local fields at protrusions during poling led to breakdown and conduction problems for thinner films. Ideally we want to go to film thicknesses of 10 nm for the 123 films in the four layer symmetric FET structure with opposing gates; the current surface roughness prevents us from achieving this with layers of less than approximately 100nm. We have improved the results but they are not, as of now, as good as desired -- only approximately one bilayer in five can withstand the application of 10V for 10 sec w/out catastrophic failure.

A method of blocking the direct deposition path (ballistic path that the larger ablated particles follow) to only allow deposition from the diffusing plasma of the plume has been implemented as a solution to this problem. In the system at Xerox this solution was only a partial help due to the physical construction of the system and the laser spot scanning system.

#### **b) Investigation of Polarization of the $\text{BaTiO}_3$**

Very precise x-ray measurements did not show any measurable dimensional changes in the films after poling so Sawyer Tower bridge polarization measurements were deemed to be the next task in pinning down the origins of the effect.

Problems with leakage in the ablated BTO films hampered accurate Sawyer-Tower measurements although some degree of hysteresis in the Capacitance vs Voltage curves has been established.

Once again smoother films would be of considerable help in reducing leakage and high local fields at protrusions (boulders).

**c) Continuation to a Symmetric Four Layer Opposed Gate Structure**

Once the surface roughness issues are resolved, regardless of the resolution of the origins of the poling effect, the fabrication and testing of a symmetric opposed gate ferroelectric insulator structure should be pursued.

**III. References**

- 1) X.X. Xi, Q. Li, C. Doughty, C. Kwon, S. Bhattacharya, A.T. Findikoglu and T. Venkatesan, "Electric Field Effect in High  $T_c$  Superconducting Ultrathin  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  Films." *Appl. Phys. Lett.*, **59** (1991) 3470-72.
- 2) X.X. Xi, C. Doughty, A. Walkenhorst, C. Kwon, Q. Li and T. Venkatesan, "Effects of Field-Induced Hole-Density Modulation on Normal State and Superconducting Transport in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ ." *Phys. Rev. Lett.*, **68** (1992) 1240-43.
- 3) G.L. Larkins et.al., "Shifts in  $T_c$  of Multilayer Ferroelectric--High  $T_c$  Structures with Poled Ferroelectric Layers," *Applied Superconductivity*, ed. by H.C. Freyhardt, *Deutsche Gessellschaft fuer Materialkunde*, **2**(1993) 1637-40.
- 4) Grover L. Larkins, Jr. and Miriam Y. Avello, "Multilayer Ferroelectric -- High  $T_c$  Structure with Poled Ferroelectric Layers", *IEEE Transactions on Applied Superconductivity*, **5**(1995) 3049-3052.
- 5) Miriam Y. Avello, "Fabrication of a two-terminal superconducting device with a poled ferroelectric control layer," Master's Thesis, Florida International University, Miami, Fla., 1996.